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Abstract

**THE SOL-GEL PROCESS FOR THE REMOTE PREPARATION
AND FABRICATION OF RECYCLE FUELS**

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The sol-gel process converts the nitrate solution products from solvent-extraction processing of spent reactor fuels to fuel materials suitable for use in remote fuel fabrication in four simple and economical steps. These steps are: denitration to obtain a reactive oxide; dispersion of this oxide in dilute nitric acid to form a hydrosol; gelation by evaporation or extraction of water from the sol; and calcination to obtain the final ceramic form desired. The process can produce either the oxides or carbides of thorium, uranium, or plutonium. It can produce powder suitable for vibratory packing into metal tubes, or microspheres for subsequent coating and use in high-temperature gas-cooled reactors. Because of this flexibility, the sol-gel process promises to become an almost universal method of preparing ceramic reactor fuel materials.

The most advanced application in our development program is the preparation of mixed thorium/uranium-233 oxide for loading into fuel tubes by vibratory compaction. A ton of this fuel has been prepared and loaded into 1000 zircaloy tubes, 40-in. (101.6 cm) long and 1/2-in. (1.27 cm) diameter, in the Kilrod Facility at Oak Ridge National Laboratory. This facility includes shielded and remotely operable equipment for the preparation of dense particles of mixed ThO_2/UO_2 by the sol-gel process, conditioning of the powder by grinding and sorting by size vibratory-compaction loading of the Zircaloy tubes, and welding and inspection of the completed element. After correction of initial mechanical difficulties, operation at 90% of the design capacity was routinely

attained with less than 3% of the elements requiring recycle because of density defects. This facility, which is our first remotely operable, integrated fuel preparation and fabrication facility, is the forerunner of the Thorium-Uranium Fuel Cycle Facility now being designed for future development of integrated fuel recycle processes.

From analysis of the radiation levels observed in the Kilorod Facility, we conclude that on a 10-kg batch scale, reactor fuel rods containing U-233 contaminated with about 500 parts/million of U-232 can be safely fabricated by use of the shadow shielding provided by about a 2-in. thickness of lead. However, final assembly of the fuel bundle, larger-scale operation, or higher levels of U-232 contamination will require complete shielding and remote operation.

An irradiation testing program is being conducted on vibratorily compacted, sol-gel thorium oxide fuels. Our findings indicate that this is a suitable fuel form for power reactor application. Irradiations to 35 000 MWd/ton at peak heat ratings of an/kd-l of 40 watts/cm have produced insignificant changes in the oxide. In capsules now being irradiated, burnups have reached 75 000 MWd/ton. At heat rating of an/kd-l of 60 watts/cm, recrystallization and sintering of the oxide are observed, but with no noticeable deleterious effects. Fission-gas release from the sol-gel oxide is comparable to that from fused oxide.

By introducing fine-particle-size carbon into the oxide sol, a mixed oxide-carbon gel can be prepared. After drying, these gels can be reacted in argon at about 1750C to give high-purity carbide. Mixed thorium-uranium dicarbide has been prepared in this manner with a density of 8.9 g/cm³ and sufficient purity for use as a reactor fuel. For the preparation of UO₂ or UC₂, it is necessary to keep the uranium mostly in the tetravalent state. Uranium dioxide sols satisfactory for the process have been prepared in two ways: by precipitation of U(OH)₄ from uranous nitrate solutions followed by aging to obtain a concentrated sol, and by electromigration of the uranyl ion into the cathode compartment of an ion-exchange membrane cell containing very dilute nitric acid. At the cathode, the uranium is reduced to the tetravalent state and precipitates to produce a sol that can be concentrated by evaporation.